

# Search for $\beta^+$ EC and ECEC processes in $^{112}\text{Sn}$ and $\beta^-\beta^-$ decay of $^{124}\text{Sn}$ to the excited states of $^{124}\text{Te}$

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## Abstract

Limits on  $\beta^+$ EC and ECEC processes in  $^{112}\text{Sn}$  and on  $\beta^-\beta^-$  decay of  $^{124}\text{Sn}$  to the excited states of  $^{124}\text{Te}$  have been obtained using a 380 cm<sup>3</sup> HPGe detector and an external source consisting of natural tin. A limit with 90% C.L. on the  $^{112}\text{Sn}$  half-life of  $0.92 \times 10^{20}$  y for the ECEC( $0\nu$ ) transition to the  $0_3^+$  excited state in  $^{112}\text{Cd}$  (1871.0 keV) has been established. This transition is discussed in the context of a possible enhancement of the decay rate by several orders of magnitude given that the ECEC( $0\nu$ ) process is nearly degenerate with an excited state in the daughter nuclide. Prospects for investigating such a process in future experiments are discussed. The  $\beta^-\beta^-$  decay limits for  $^{124}\text{Sn}$  to the excited states of  $^{124}\text{Te}$  were obtained on the level of  $(0.8 - 1.2) \times 10^{21}$  y at the 90% C.L.

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*Key words:* double-beta decay, double electron capture,  $^{112}\text{Sn}$ ,  $^{124}\text{Sn}$ .

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## 1 Introduction

Interest in neutrinoless double-beta decay has seen a significant renewal in recent years after evidence for neutrino oscillations was obtained from the results of atmospheric, solar, reactor and accelerator neutrino experiments (see, for example, the discussions in [1,2,3]). These results are impressive proof that neutrinos have a non-zero mass. However, the experiments studying neutrino oscillations are not sensitive to the nature of the neutrino mass (Dirac or Majorana) and provide no information on the absolute scale of the neutrino masses, since such experiments are sensitive only to the difference of the masses,  $\Delta m^2$ . The detection and study of  $0\nu\beta\beta$  decay may clarify the following problems of neutrino physics (see discussions in [4,5,6]): (i) neutrino nature: whether the neutrino is a Dirac or a Majorana particle, (ii) absolute neutrino mass scale (a measurement or a limit on  $m_1$ ), (iii) the type of neutrino mass hierarchy (normal, inverted, or quasidegenerate), (iv) CP violation in the lepton sector (measurement of the Majorana CP-violating phases). At the present time only limits on the level of  $\sim 10^{24} - 10^{25}$  y for half-lives and  $\sim 0.3 - 1$  eV for effective Majorana neutrino mass  $\langle m_\nu \rangle$  have been obtained in the best modern experiments (see recent reviews [7,8,9]).

In connection with the  $0\nu\beta\beta$  decay, the detection of double beta decay with emission of two neutrinos ( $2\nu\beta\beta$ ), which is an allowed process of second order in the Standard Model, provides the possibility for experimental determination of the nuclear matrix elements (NME) involved in the double beta decay processes. This leads to the development of theoretical schemes for nuclear matrix-element calculations both in connection with the  $2\nu\beta\beta$  decays as well as the  $0\nu\beta\beta$  decays ([10,11,12]). At present,  $2\nu\beta\beta$  to the ground state of the final daughter nucleus has been measured for ten nuclei (a review of results is given in Ref. [13]).

Recently, it has been pointed out that the  $2\nu\beta\beta$  decay allows one to investigate particle properties, in particular whether the Pauli exclusion principle is violated for neutrinos and thus neutrinos at least partially obey Bose-Einstein statistics [14,15].

The  $\beta\beta$  decay can proceed through transitions to the ground state as well as to various excited states of the daughter nucleus. Studies of the latter transitions provide supplementary information about  $\beta\beta$  decay.

As it was shown in Ref. [16], by using low-background facilities utilizing High Purity Germanium (HPGe) detectors, the  $2\nu\beta\beta$  decay to the  $0_1^+$  level in the daughter nucleus may be detected. Soon after this double beta decay of  $^{100}\text{Mo}$  to the  $0^+$  excited state at 1130.29 keV in  $^{100}\text{Ru}$  was observed [17]. This result was confirmed in independent experiments [18,19,20]. In 2004 for the first time

the transition was detected in  $^{150}\text{Nd}$  [21]. Recently additional isotopes, have become of interest in studies of  $2\nu\beta\beta$  decay to the  $0_1^+$  level [22,23,24].

The  $0\nu\beta\beta$  transition to excited states of daughter nuclei has a clear signature for such decays. It is worthy of a special note here that in addition to two electrons with a fixed total energy, there are one ( $0^+ - 2^+$  transition) or two ( $0^+ - 0^+$  transition) photons with their energies being strictly fixed. In a hypothetical experiment which detects all the decay products with a high efficiency and a high energy resolution the background can be reduced to nearly zero. It is possible that this idea will be used in future experiments featuring a large mass of the isotope under study (as it was mentioned in [21,22,25]). In Ref. [26] it was stated that detection of this transition can distinguish between the  $0\nu\beta\beta$  mechanisms (light and heavy Majorana neutrino exchange mechanisms, trilinear R-parity breaking mechanisms etc.). So the search for  $\beta\beta$  transitions to excited states has its own special interest.

Most double beta decay investigations have concentrated on the  $\beta^-\beta^-$  decay. Much less attention has been given to the investigation of  $\beta^+\beta^+$ ,  $\beta^+\text{EC}$  and  $\text{ECEC}$  processes (here EC denotes electron capture). There are 34 candidates for these processes. Only six nuclei can undergo all of the above mentioned processes and 16 nuclei can undergo  $\beta^+\text{EC}$  and  $\text{ECEC}$  while 12 can undergo only  $\text{ECEC}$ . Detection of the neutrinoless mode in the above processes enable one to determine the effective Majorana neutrino mass  $\langle m_\nu \rangle$  and parameters of right-handed current admixture in electroweak interaction ( $\langle \lambda \rangle$  and  $\langle \eta \rangle$ ). Detection of the two-neutrino mode in the above processes lets one determine the magnitude of the nuclear matrix elements involved, which is very important in view of the theoretical calculations for both the  $2\nu$  and the  $0\nu$  modes of double beta decay. Interestingly, it was demonstrated in Ref. [27] that if the  $\beta^-\beta^-(0\nu)$  decay is detected, then the experimental limits on the  $\beta^+\text{EC}(0\nu)$  half-lives can be used to obtain information about the relative importance of the Majorana neutrino mass and right-handed current admixtures in electroweak interactions.

The  $\beta^+\beta^+$  and  $\beta^+\text{EC}$  processes are less favorable due to smaller kinetic energy available for the emitted particles and Coulomb barrier for the positrons. However, an attractive feature of these processes from the experimental point of view is the possibility of detecting either the coincidence signals from four (two) annihilation  $\gamma$ -rays and two (one) positrons, or the annihilation  $\gamma$ -rays only. It is difficult to investigate the  $\text{ECEC}$  process because one only detects the low energy X-rays. It is also interesting to search for transitions to the excited states of daughter nuclei, which are easier to detect given the cascade of higher energy gammas [28]. In Ref. [29] it was the first mentioned that in the case of  $\text{ECEC}(0\nu)$  transition a resonance condition can exist for transition to the "right energy" of the excited level for the daughter nucleus, here the decay energy is close to zero. In 1982 the same idea was proposed for the transition to

the ground state [30]. In 1983 this possibility was discussed for the transition  $^{112}\text{Sn}$  to  $^{112}\text{Cd}$  ( $0^+$ ; 1871 keV) [31]. In 2004 the idea was reanalyzed in Ref. [32] and new resonance conditions for the decay were formulated. The possible enhancement of the transition rate was estimated as  $\sim 10^6$  [31,32]. This means that this process starts to be competitive with  $0\nu\beta\beta$  decay for the neutrino mass sensitivity and is interesting to check experimentally. There are several candidates for which resonance transition, to the ground ( $^{152}\text{Gd}$ ,  $^{164}\text{Eu}$  and  $^{180}\text{W}$ ) and to the excited states ( $^{74}\text{Se}$ ,  $^{78}\text{Kr}$ ,  $^{96}\text{Ru}$ ,  $^{106}\text{Cd}$ ,  $^{112}\text{Sn}$ ,  $^{130}\text{Ba}$ ,  $^{136}\text{Ce}$  and  $^{162}\text{Er}$ ) of daughter nuclei exist [24,32]. The precision needed to realize resonance condition is well below 1 keV. To select the best candidate from the above list one will have to know the atomic mass difference with an accuracy better than 1 keV and such measurements are planned for the future. Recently the experimental search for such a resonance transition in  $^{74}\text{Se}$  to  $^{74}\text{Ge}$  ( $2^+$ ; 1206.9 keV) was performed yielding a limit  $T_{1/2} > 5.5 \times 10^{18}$  y [33]. Very recently  $^{112}\text{Sn}$  was investigated [34,35] and a limit of  $T_{1/2} > 1.6 \times 10^{18}$  y was obtained for the transition to the  $0^+$  state at 1871 keV [35].

In this article the results of an experimental investigation of the  $\beta^+\text{EC}$  and ECEC processes in  $^{112}\text{Sn}$  and  $\beta^-\beta^-$  decay of  $^{124}\text{Sn}$  to the excited states of  $^{124}\text{Te}$  are presented.

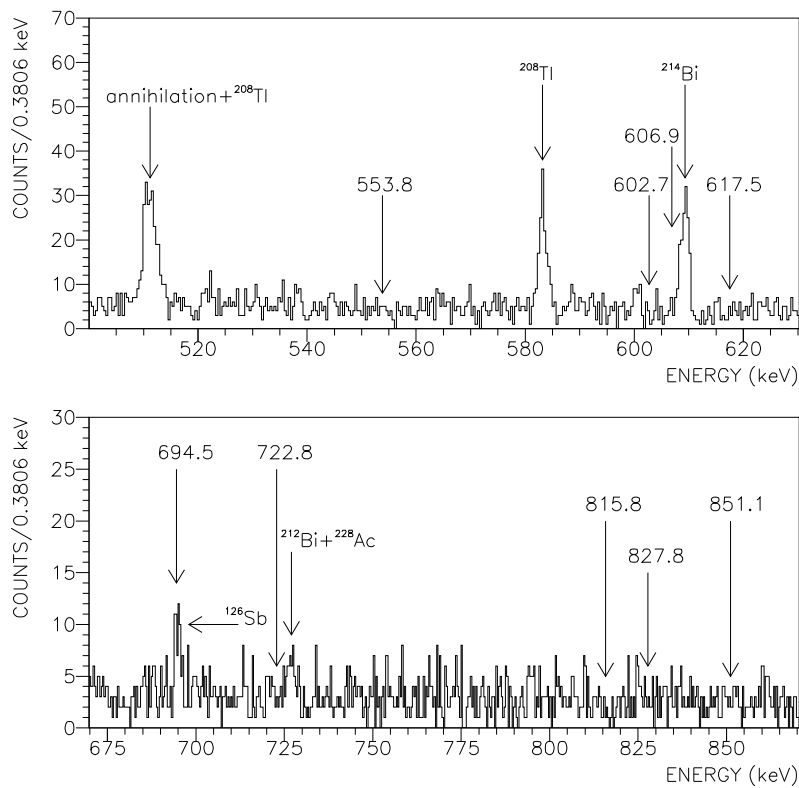


Fig. 1. Energy spectrum with 3975.3 g of natural Sn for 2385.4 h of measurement in the ranges investigated ([500-630] and [670-870] keV).

## 2 Experimental

The experiment was performed in the Modane Underground Laboratory at a depth of 4800 m w.e.. The natural tin sample, guaranteed to be 99.99 % pure, was measured using a 380 cm<sup>3</sup> low-background HPGe detector.

The HPGe spectrometer is a p-type crystal with the cryostat, endcap and majority of the mechanical components made of a very pure Al-Si alloy. The cryostat has a J-type geometry to shield the crystal from radioactive impurities in the dewar. The passive shielding consisted of 4 cm of Roman-era lead and 3-10 cm of OFHC copper inside 15 cm of ordinary lead. To remove <sup>222</sup>Rn gas, one of the main sources of the background, a special effort was made to minimize the free space near the detector. In addition, the passive shielding was enclosed in an aluminum box flushed with radon free air (< 10 mBq/m<sup>3</sup>) delivered by a radon free factory installed in the Modane Underground Laboratory.

The electronics consisted of currently available spectrometric amplifiers and an 8192 channel ADC. The energy calibration was adjusted to cover the energy range from 50 keV to 3.5 MeV, and the energy resolution was 2.0 keV for the 1332-keV line of <sup>60</sup>Co. The electronics were stable during the experiment due to the constant conditions in the laboratory (temperature of  $\approx 23$  °C, hygrometric degree of  $\approx 50\%$ ). A daily check on the apparatus assured that the counting rate was statistically constant.

The first portion of natural tin (2543.2 g) was placed in a derlin Marinelli box surrounding the HPGe detector. The second portion (1432.1 g) was placed on the endcap of the HPGe detector. The total mass of tin was 3975.3 g, 240.24 g was <sup>124</sup>Sn (natural abundance is 5.79%) and 36.35 g was <sup>112</sup>Sn (natural abundance is 0.97%). The duration of the measurement was 2385.43 hours.

The natural tin sample was found to have a small quantity of <sup>40</sup>K ((3.8  $\pm$  0.6) mBq/kg) and also cosmogenic and natural tin radioactivities, i.e. the 158-keV decreasing peak of <sup>117m</sup>Sn (13.76 d), the 391-keV decreasing peak of <sup>113</sup>Sn (115.09 d), (46  $\pm$  9)  $\mu$ Bq/kg of <sup>126</sup>Sn (10<sup>5</sup> y), and (159  $\pm$  48)  $\mu$ Bq/kg of <sup>125</sup>Sb (2.75856 y). The natural radioactivities had only limits which were < 94  $\mu$ Bq/kg of <sup>228</sup>Ac, < 95  $\mu$ Bq/kg of <sup>226</sup>Ra, < 37  $\mu$ Bq/kg of <sup>137</sup>Cs, and < 20  $\mu$ Bq/kg of <sup>60</sup>Co.

The search for different  $\beta^+$ EC and ECEC processes in <sup>112</sup>Sn and  $\beta^-\beta^-$  decay of <sup>124</sup>Sn to the excited states of <sup>124</sup>Te were carried out using the germanium detector to look for  $\gamma$ -ray lines corresponding to these processes. Gamma-ray spectra of selected energy ranges are shown in Fig. 1-3. These spectra correspond to regions-of-interest for the different decay modes of <sup>112</sup>Sn and <sup>124</sup>Sn.

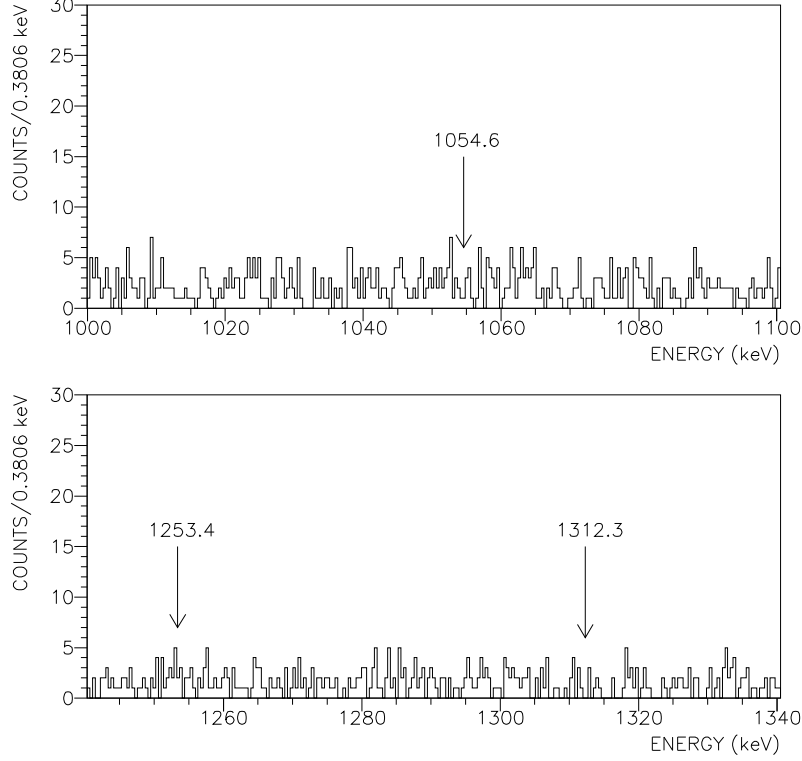


Fig. 2. Energy spectrum with 3975.3 g of natural Sn for 2385.4 h of measurement in the ranges investigated ([1000-1100] and [1240-1340] keV).

### 3 Search for $\beta^+EC$ and ECEC processes in $^{112}\text{Sn}$

The decay scheme for the triplet  $^{112}\text{Cd}$ - $^{112}\text{In}$ - $^{112}\text{Sn}$  is shown in Fig. 4 [36,37]. The  $\Delta M$  (difference of parent and daughter atomic masses) value of the transition is  $1919.5 \pm 4.8$  keV [38] and the natural abundance of  $^{112}\text{Sn}$  is 0.97%. The following decay processes are possible:

$$e_b^- + (A, Z) \rightarrow (A, Z - 2) + e^+ + X \quad (\beta^+EC; 0\nu) \quad (1)$$

$$e_b^- + (A, Z) \rightarrow (A, Z - 2) + e^+ + 2\nu + X \quad (\beta^+EC; 2\nu) \quad (2)$$

$$2e_b^- + (A, Z) \rightarrow (A, Z - 2) + 2X \quad (ECEC; 0\nu) \quad (3)$$

$$2e_b^- + (A, Z) \rightarrow (A, Z - 2) + 2\nu + 2X \quad (ECEC; 2\nu) \quad (4)$$

where  $e_b$  is an atomic electron and  $X$  represents X-rays or Auger electrons. Introduced here is the notation  $Q'$  which is the effective  $Q$ -value defined as  $Q' = \Delta M - \epsilon_1 - \epsilon_2$  for the ECEC transition and  $Q' = \Delta M - \epsilon_1 - 2m_e c^2$  for the  $\beta^+EC$  process;  $\epsilon_i$  is the electron binding energy of a daughter nuclide. For  $^{112}\text{Cd}$ ,  $\epsilon$  is equal to 26.7 keV for the K shell and 4.01 keV, 3.72 keV and 3.54 keV for the L shell (2s,  $2p_{1/2}$  and  $2p_{3/2}$  levels). In the case of the L shell the

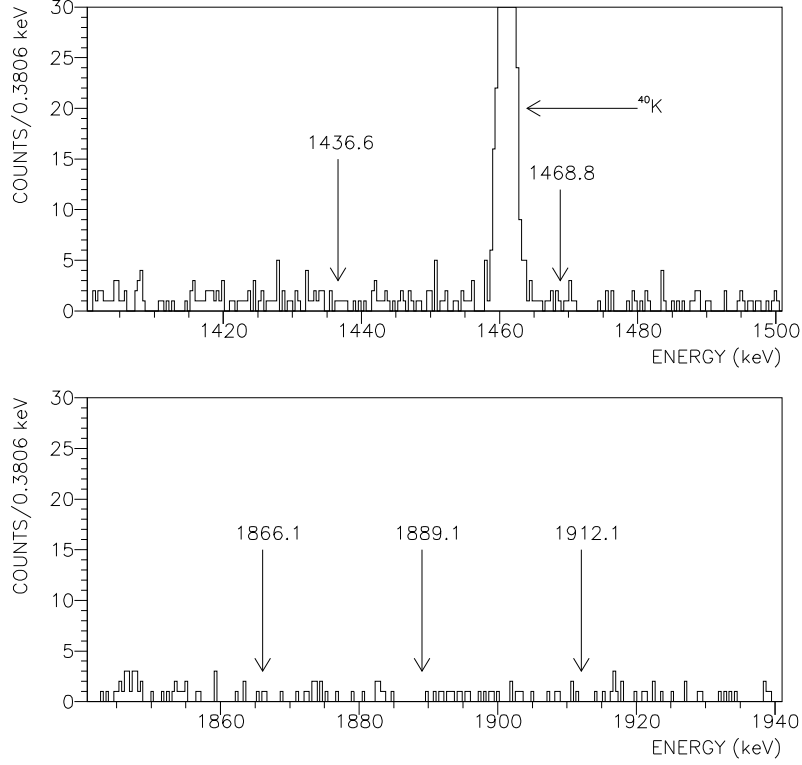


Fig. 3. Energy spectrum with 3975.3 g of natural Sn for 2385.4 h of measurement in the ranges investigated ([1400-1500] and [1850-1940] keV).

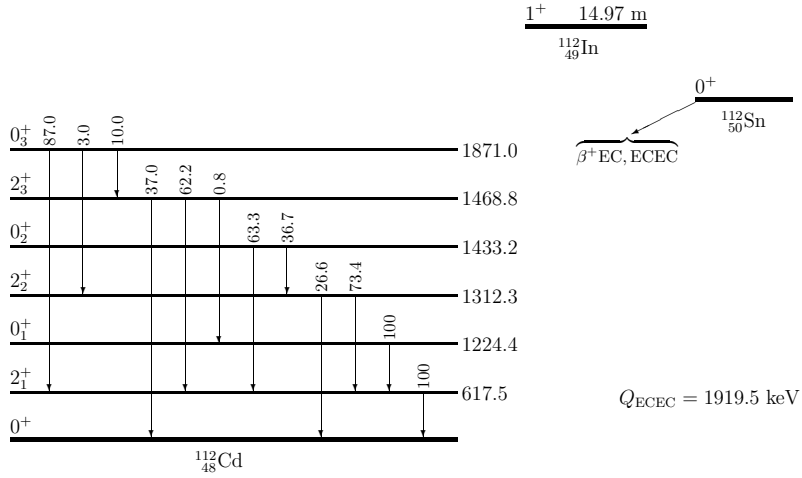


Fig. 4. Decay scheme of  $^{112}\text{Sn}$ . Only the investigated levels associated with  $\gamma$ -rays are shown. Transition probabilities are given in percents.

resolution of the HPGe detector prohibits separation of the lines so we center the study on the 3.72 keV line.

Investigations were made of the  $\beta^+\text{EC}$  transitions to the ground and the  $2_1^+$  excited states. Additionally, the ECEC transitions to the ground state and six excited states ( $2_1^+$ ,  $0_1^+$ ,  $2_2^+$ ,  $0_2^+$ ,  $2_3^+$  and  $0_3^+$ ) were investigated.

### 3.1 ECEC transitions

The ECEC( $0\nu + 2\nu$ ) transition to the excited states of  $^{112}\text{Cd}$  is accompanied with  $\gamma$ -quanta with different energies (see decay scheme in Fig. 4). These  $\gamma$ -quanta were used in the search. The approach is not sensitive to ECEC( $2\nu$ ) to the ground state because X-rays are absorbed in the sample and cannot reach the sensitive volume of the HPGe detector.

The ECEC( $0\nu$ ) transition to the ground state of the daughter nuclei was considered for three different electron capture cases:

- 1) Two electrons are captured from the L shell. In this case,  $Q'$  is equal to  $1912.1 \pm 4.8$  keV and the transition is accompanied by a bremsstrahlung  $\gamma$ -quantum with an energy  $\sim 1912.1$  keV.
- 2) One electron is captured from the K shell, another from the L shell. In this case,  $Q'$  is equal to  $1889.1 \pm 4.8$  keV and the transition is accompanied by a bremsstrahlung  $\gamma$ -quantum with an energy  $\sim 1889.1$  keV.
- 3) Two electrons are captured from the K shell. In this case,  $Q'$  is equal to  $1866.1 \pm 4.8$  keV and the transition is accompanied by  $\gamma$ -quantum with an energy  $\sim 1866.1$  keV. In fact this transition is strongly suppressed (forbidden) because of momentum conservation. So in this case the more probable outcome is the emission of  $e^+e^-$  pair [40] which gives two annihilation  $\gamma$ -quanta with an energy of 511 keV.

The Bayesian approach [41] was used to estimate limits on transitions of  $^{112}\text{Sn}$  to the ground and excited states of  $^{112}\text{Cd}$ . To construct the likelihood function, every bin of the spectrum is assumed to have a Poisson distribution with its mean  $\mu_i$  and the number of events equal to the content of the  $i$ th bin. The mean can be written in the general form,

$$\mu_i = N \sum_m \varepsilon_m a_{mi} + \sum_k P_k a_{ki} + b_i. \quad (5)$$

The first term in (5) describes the contribution of the investigated process that may have a few  $\gamma$ -lines contributing appreciably to the  $i$ th bin. The parameter  $N$  is the number of decays,  $\varepsilon_m$  is the detection efficiency of the  $m$ th  $\gamma$ -line and  $a_{mi}$  is the contribution of the  $m$ th line to the  $i$ th bin. For low-background measurements a  $\gamma$ -line may be taken to have a gaussian shape. The second term gives contributions of background  $\gamma$ -lines. Here  $P_k$  is the area of the  $k$ th  $\gamma$ -line and  $a_{ki}$  is its contribution to the  $i$ th bin. The third term represents the so-called “continuous background” ( $b_i$ ), which has been selected as a straight-line fit after rejecting all peaks in the region-of-interest. We have selected this region as the peak to be investigated  $\pm 30$  standard deviations ( $\approx 20$



keV). The likelihood function is the product of probabilities for selected bins. Normalizing over the parameter  $N$  gives the probability density function for  $N$ , which is used to calculate limits for  $N$ . To take into account errors in the  $\gamma$ -line shape parameters, peak areas, and other factors, one should multiply the likelihood function by the error probability distributions for these values and integrate, to provide the average probability density function for  $N$ .

In the case of the ECEC( $0\nu$ ) transition to the ground state of  $^{112}\text{Cd}$  there is a large uncertainty in the energy of the bremsstrahlung  $\gamma$ -quantum because of a poor accuracy in  $\Delta M$  ( $\pm 4.8$  keV). Thus the position of the peak was varied in the region of the uncertainty and the most conservative value of the limit for the half-life was selected.

The photon detection efficiency for each investigated process has been computed with the CERN Monte Carlo code GEANT 3.21. Special calibration measurements with radioactive sources and powders containing well-known  $^{226}\text{Ra}$  activities confirmed that the accuracy of these efficiencies is about 10%.

The final results are presented in Table 1. In the 4-th column are the best previous experimental results from Ref. [34,35] for comparison. In the last column, the theoretical estimations for ECEC( $2\nu$ ) transitions obtained under the assumption of single intermediate nuclear state dominance are also presented [42].

Concerning the ECEC( $0\nu$ ) processes, the plan is to observe a resonant transition to the 1871.0 keV excited state of  $^{112}\text{Cd}$ . In this case we look for two peaks, at 617.5 keV and 1253.4 keV. In fact, the experimental spectrum has some excess of events in the range of 1253.4 keV,  $\sim 2.3\sigma$  above the continuous background. At the same time there are no extra events in the energy range of 617.5 keV. The total effect (taking into account both peaks) is only  $1.5\sigma$ . If all the extra events are connected with the ECEC( $0\nu$ ) transition of  $^{112}\text{Sn}$  to the 1871.0 keV excited state of  $^{112}\text{Cd}$  then the corresponding half-life value for this process is at the level of  $\sim 10^{20}$  y. The conservative approach gives the limit  $T_{1/2} > 0.92 \times 10^{20}$  y at the 90% C.L.

### 3.2 $\beta^+EC$ transitions

The  $\beta^+EC(0\nu + 2\nu)$  transition to the ground state is accompanied by two annihilation  $\gamma$ -quanta with an energy of 511 keV. These  $\gamma$ -quantum were used to search for this transition. In the case of the  $\beta^+EC(0\nu + 2\nu)$  transition to the  $2_1^+$  excited state the 617.4 keV  $\gamma$ -quanta was also detected. To obtain limits on these transitions the analysis described in Section 3.1 was used. Again the photon detection efficiencies for each investigated process was computed with the CERN Monte Carlo code GEANT 3.21. and are presented in Table 1. In

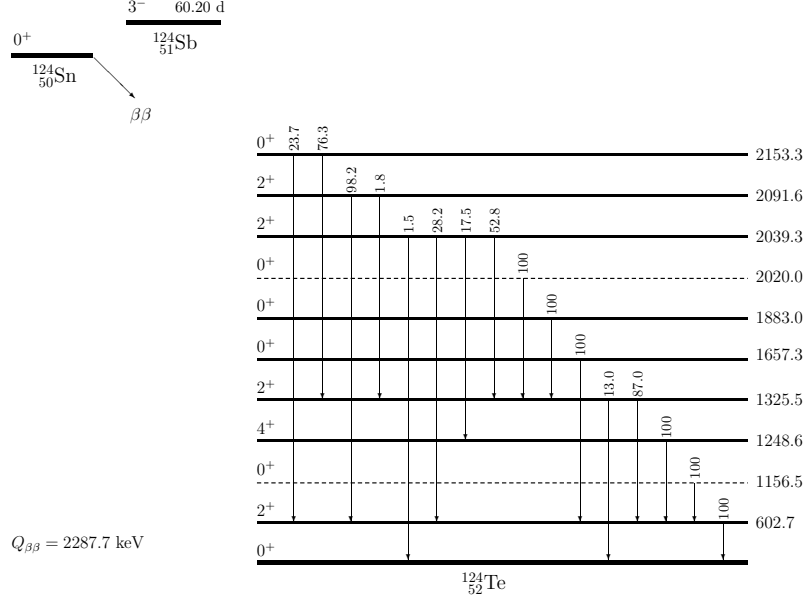


Fig. 5. Decay scheme of  $^{124}\text{Sn}$  are taken from [43] and two levels (1156.5 keV and 2020.0 keV) are added from [44]. The investigated levels are associated with the  $\gamma$ -rays shown. Transition probabilities are given in percents.

the last two columns are the best previous results and theoretical predictions for comparison.

#### 4 Search for $\beta\beta$ decay of $^{124}\text{Sn}$ to the excited states of $^{124}\text{Te}$

The decay scheme for the triplet  $^{124}\text{Sn}$ - $^{124}\text{Sb}$ - $^{124}\text{Te}$  is shown in Fig. 5 [37,43,44]. The  $\Delta M$  in this case is  $2287.7 \pm 2.1$  keV [38]. The following processes are possible:

$$(A, Z) \rightarrow (A, Z + 2) + 2e^- + 2\tilde{\nu} \quad (2\nu\beta\beta) \quad (6)$$

$$(A, Z) \rightarrow (A, Z + 2) + 2e^- \quad (0\nu\beta\beta) \quad (7)$$

$$(A, Z) \rightarrow (A, Z + 2) + 2e^- + \chi^0 \quad (0\nu\chi^0\beta\beta) \quad (8)$$

We will consider only transitions to the excited states of  $^{124}\text{Te}$ . The most intensive  $\gamma$ -rays from the decay scheme (Fig. 5) were used for analysis. The detection efficiencies for photopeaks corresponding to specified  $\gamma$ -quanta are given in Table 2. The efficiencies were again computed with the CERN Monte Carlo code GEANT 3.21.

The spectra for specified energy ranges are shown in Fig. 1-3. There are no statistically significant peaks at the indicated energies. The lower half-life lim-

Table 1

The experimental limits and theoretical predictions for the  $\beta^+$ EC and ECEC processes in  $^{112}\text{Sn}$ . \*) For transition with irradiation of  $e^+e^-$  pair - see text.

Transition	Energy of $\gamma$ -rays, keV (Efficiency)	$T_{1/2}^{exp}, 10^{20} \text{ y (C.L. 90\%)}$		$T_{1/2}^{th}(2\nu), \text{ y}$ [42]
		Present work	Previous works	
$\beta^+$ EC( $0\nu + 2\nu$ ); g.s.	511.0 (5.05 %)	0.12	0.0091 [34] 0.041 [35]	$3.8 \times 10^{24}$
$\beta^+$ EC( $0\nu + 2\nu$ ); $2_1^+$	617.5 (1.85 %)	0.94	0.014 [35]	$2.3 \times 10^{32}$
ECEC( $0\nu$ ) $L^1L^2$ ; g.s.	1912.1 (1.44 %)	1.3	-	
ECEC( $0\nu$ ) $K^1L^2$ ; g.s.	1889.1 (1.45 %)	1.8	0.0099 [35]	
ECEC( $0\nu$ ) $K^1K^2$ ; g.s.	1866.1 (1.47 %)	1.3	-	
	511.0 (5.05 %)	0.12*)	-	
ECEC( $0\nu$ ); $2_1^+$	617.5 (2.11 %)	1.1	0.014 [35]	
ECEC( $0\nu$ ); $0_1^+$	606.9 (1.90 %)	1.2	0.014 [35]	
	617.5 (1.87 %)			
ECEC( $0\nu$ ); $2_2^+$	617.5 (1.37 %)	0.89	0.014 [35]	
	1312.3 (0.46 %)			
ECEC( $0\nu$ ); $0_2^+$	617.5 (1.73 %)	1.6	0.014 [35]	
	815.8 (1.08 %)			
ECEC( $0\nu$ ); $2_3^+$	617.5 (1.20 %)	0.93	0.014 [35]	
	851.1 (1.06 %)			
	1468.8 (0.58 %)			
ECEC( $0\nu$ ); $0_3^+$	617.5 (2.00 %)	0.92	0.016 [35]	
	1253.4 (1.39 %)			
ECEC( $2\nu$ ); $2_1^+$	617.5 (2.41 %)	1.2	0.014 [35]	$4.9 \times 10^{28}$
ECEC( $2\nu$ ); $0_1^+$	606.9 (2.11 %)	1.4	0.014 [35]	$7.4 \times 10^{24}$
	617.5 (2.09 %)			
ECEC( $2\nu$ ); $2_2^+$	617.5 (1.55 %)	1.0	0.014 [35]	$1.9 \times 10^{32}$
	1312.3 (0.51 %)			

Table 1  
Continued.

Transition	Energy of $\gamma$ -rays, keV (Efficiency)	$T_{1/2}^{exp}, 10^{20}$ y (C.L. 90%)		$T_{1/2}^{th}(2\nu)$ , y [42]
		Present work	Previous works	
ECEC( $2\nu$ ); $0_2^+$	617.5 (1.88 %)	1.8	0.014 [35]	-
	815.8 (1.21 %)			
ECEC( $2\nu$ ); $2_3^+$	617.5 (1.32 %)	1.0	0.014 [35]	$6.2 \times 10^{31}$
	851.1 (1.19 %)			
	1468.8 (0.64 %)			
ECEC( $2\nu$ ); $0_3^+$	617.5 (2.00 %)	0.92	0.016 [35]	$5.4 \times 10^{34}$
	1253.4 (1.39 %)			

its reported in Table 2 have been calculated using the likelihood function described in Section 3.1. Available data on  $\beta\beta$  decay of  $^{124}\text{Sn}$  from other experimental works and theoretical estimates are presented in Table 2.

Our limits on  $\beta\beta$  transitions of  $^{124}\text{Sn}$  to the excited states of daughter nucleus are valid for the  $0\nu$ ,  $2\nu$  modes and all types of decay with Majoron emission ( $0\nu\chi^0$ ).

## 5 Discussion

Limits obtained for the  $\beta^+\text{EC}$  and ECEC processes in  $^{112}\text{Sn}$  are on the level of  $\sim (0.1 - 2) \times 10^{20}$  y or  $\sim 10$ -200 times better than the best previous results [34,35] (see Table 1). As one can see from Table 1 the theoretical predictions for  $2\nu$  transitions are much higher than the measured limits. The sensitivity of such experiments can still be increased with the experimental possibilities being the following:

- 1) Given 4 kg of enriched  $^{112}\text{Sn}$  in the setup described in Section 2, the sensitivity after one year of measurement will be  $\sim 3 \cdot 10^{22}$  y.
- 2) With 200 kg of enriched  $^{112}\text{Sn}$  using an installation such as GERDA [47] or MAJORANA [48,49] where 500-1000 kg of low-background HPGe detectors are planned.<sup>2</sup> The sensitivity after 10 years of measurement may reach  $\sim 10^{26}$  y. Thus there is a chance of detecting the  $\beta^+\text{EC}(2\nu)$  transition of  $^{112}\text{Sn}$  to the

<sup>2</sup> Imaging around each of the  $\sim 200$  HPGe crystals can be  $\sim 1$  kg of very pure  $^{112}\text{Sn}$ . Both  $^{76}\text{Ge}$  and  $^{112}\text{Sn}$  will be investigated at the same time.

Table 2

Theoretical and experimental results for  $(0\nu + 2\nu + 0\nu\chi^0)\beta\beta$  decay of  $^{124}\text{Sn}$  to the excited states of  $^{124}\text{Te}$ . Limits are given at the 90% C.L.

Excited state, (keV)	Energy of $\gamma$ -rays, keV (Efficiency)	$(T_{1/2}^{2\nu})^{th}$ , y [39]	$(T_{1/2}^{0\nu+2\nu+0\nu\chi^0})_{exp}$ , $10^{21}$ y	
			Present work	Previous works
$2_1^+(602.7)$	602.7 (2.40%)	$6.5 \cdot 10^{26}$	$> 0.91$	$> 0.0031$ [35]
				$> 0.0023$ [34]
				$> 0.033$ [45]
$0_1^+(1156.5)$	553.8 (2.20%)	$2.7 \cdot 10^{21}$	$> 1.1$	$> 0.0077$ [35]
	602.7 (2.15%)			$> 0.0067$ [34]
$2_2^+(1325.5)$	602.7 (1.86%)	$1.7 \cdot 10^{27}$	$> 0.94$	$> 0.0044$ [35]
	722.8 (1.76%)			$> 0.0079$ [34]
$0_2^+(1657.3)$	602.7 (2.14%)	—	$> 1.2$	$> 0.0079$ [35]
	1054.6 (1.72%)			
$0_3^+(1882.98)$	557.5 (1.96%)	—	$> 1.2$	—
	602.7 (1.65%)			
	722.8 (1.55%)			
$0_4^+(2020.0)$	602.7 (1.64%)	—	$> 0.82$	$> 0.0044$ [35]
	694.5 (1.81%)			
	722.8 (1.53%)			
$2_3^+(2039.3)$	602.7 (1.82%)	—	$> 0.86$	$> 0.0044$ [35]
	722.8 (0.82%)			
	1436.6 (0.46%)			
$2_4^+(2091.6)$	602.7 (2.13%)	—	$> 0.96$	$> 0.0031$ [35]
	1488.9 (1.43%)			
$0_5^+(2153.3)$	602.7 (1.78%)	—	$> 0.95$	—
	722.8 (1.18%)			
	1550.6 (0.37%)			

ground state and ECEC( $2\nu$ ) transition to the  $0_1^+$  excited state (see theoretical predictions in Table 1).

In the case of the ECEC( $0\nu$ ) transition to the  $0_3^+$  (1871.0 keV) excited state of  $^{112}\text{Cd}$  a very small excess of events ( $\sim 1.5\sigma$ ) was detected. The effect is weak and only a limit can be extracted in this case. So the search for this

process continues into the future. Note that the ECEC( $2\nu$ ) transition to the  $0_3^+$  excited state is strongly suppressed because of the very small phase space volume. In contrast, the probability of the  $0\nu$  transition should be strongly enhanced if the resonance condition is realized. In Ref. [31,32] the "increasing factor" was estimated as  $\sim 10^6$  and can be even higher. Then if the "positive" effect is observed in future experiments it is the ECEC( $0\nu$ ) process. This will mean that lepton number is violated and the neutrino is a Majorana particle. To extract the  $\langle m_\nu \rangle$  value one has to know the nuclear matrix element for this transition and therefore the exact value of  $\Delta M$  (see [31,32]). The necessary accuracy for  $\Delta M$  is better than 1 keV and this is a realistic task (in Ref. [46] the possibility of measurements with accuracy  $\sim 200$  eV is discussed).

Two of different descriptions for the resonance were discussed in the past. In Ref. [31] the resonance condition is realized when  $Q'$  is close to zero. They treat the process as (1S,1S) double electron capture and  $Q'$  is equal to  $-4.9 \pm 4.8$  keV ( $1\sigma$  error). Thus there is a probability that  $Q'$  is less than 1 keV. In this case one has a few daughter-nucleus  $\gamma$  rays (see scheme in fig.1) and two Cd K X-rays, one of which may have its energy shifted by the mismatch in energies between the parent atom and the almost degenerate virtual daughter state. In Ref. [32,50] the decay is treated as (1S,2P) double electron capture with irradiation of an internal bremsstrahlung photon. The  $Q'$  value (energy of the bremsstrahlung photon) is  $18.1 \pm 4.8$  keV. The resonance condition for the transition is realized when  $E_{brems} = Q_{res} = |E(1S, Z-2) - E(2P, Z-2)|$ , i.e. when the bremsstrahlung photon energy becomes comparable to the  $2P - 1S$  atomic level difference in the final atom ( $23 \text{ keV}$ )<sup>3</sup>. Anticipated, taking into account uncertainties in the  $Q'$  value, is that the real  $Q'$  value is equal to 23 keV with an accuracy better than 1 keV and the resonance condition is realized. There are a few daughter-nucleus  $\gamma$  rays (see scheme in fig.1), one Cd K X-ray and bremsstrahlung photon with energy  $\sim K_\alpha$ . The bremsstrahlung photon may have its energy shifted by the mismatch in energy between the parent atom and the almost degenerate virtual daughter state.

Finally, both approaches predict the same experimental signature for this transition and need to know with better accuracy the value of  $\Delta M$  to be sure that the resonance condition is really valid. New theoretical investigations of this transition are needed.

Limits obtained for  $\beta\beta$  decay of  $^{124}\text{Sn}$  to the excited state of  $^{124}\text{Te}$  are on the level  $\sim (0.8 - 1.2) \times 10^{21}$  y or  $\sim 30$ -300 times better than the best previous results [34,35,45] (see Table 2). One can see from Table 2 the theoretical prediction for  $2\nu$  transitions to the  $0_1^+$  excited state is  $2.7 \cdot 10^{21}$  y and only  $\sim 2.5$  times higher than the obtained limit. Therefore there is a chance to detect

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<sup>3</sup> The same effect was theoretically predicted and then experimentally confirmed for single electron capture (see discussion in [50])

this decay in the future with more sensitive experiments. The experimental possibilities here are approximately the same as for  $^{112}\text{Sn}$  (see above):

- 1) With 4 kg of enriched  $^{124}\text{Sn}$  in the experiment described in Section 2, the sensitivity after one year of measurement will be  $\sim 3 \cdot 10^{22}$  y.
- 2) With 200 kg of enriched  $^{124}\text{Sn}$  using an installation such as GERDA [47] or MAJORANA [48,49] where 500-1000 kg of low-background HPGe detectors are planned. The sensitivity after 10 years of measurement may reach  $\sim 10^{26}$  y.

## 6 Conclusion

New limits on  $\beta^+\text{EC}$  and ECEC processes in  $^{112}\text{Sn}$  and on  $\beta^-\beta^-$  decay of  $^{124}\text{Sn}$  to the excited states of  $^{124}\text{Te}$  have been obtained using a 380 cm<sup>3</sup> HPGe detector and an external source consisting of natural tin. In addition, it has been demonstrated that, in the future larger-scale experiments, the sensitivity to the ECEC( $0\nu$ ) processes for  $^{112}\text{Sn}$  can reach the order of  $10^{26}$  y. Under resonant conditions this decay will be competitive with  $0\nu\beta\beta$  decay. The same sensitivity can be reached for double beta decay of  $^{124}\text{Sn}$  to the excited states of  $^{124}\text{Te}$ .

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